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ADVANCED DIAGNOSTICS FOR REACTING FLOWS

AFOSR 01-1-0145

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SUMMARY/OVERVIEW:

The goal of this research is to develop advanced laser-based techniques for non-intrusive measurements relevant to air-breathing combustion. In general, the program emphasizes spectrally-resolved absorption using tunable laser sources and planar laser-induced fluorescence (PLIF), conducted using either near-infrared or ultraviolet laser sources. Detailed below is progress on the exploration of IR PLIF as a diagnostic for imaging IR-active gases and on the suitability of pentanone as an alternative flow tracer to acetone for PLIF imaging. Furthermore, a novel absorption sensor for NO_2 at temperatures up to 1900 K has been developed, and continuing work is reported on the spectroscopy of high-pressure gases.

TECHNICAL DISCUSSION:

Infrared PLIF Imaging

Infrared Planar Laser-Induced Fluorescence (IR PLIF) has high potential for visualizing CO , CO_2 , and other IR-active species in reactive flows. Fig. 1 shows a basic IR PLIF set up, including an IR laser source. The slow characteristic times of vibrational relaxation and the large mole fractions for CO and CO_2 in typical flows lead to high IR PLIF signal levels, despite the low emission rates typical of vibrational transitions. Analyses of rotational energy transfer (RET) and vibrational energy transfer (VET) show that excitation schemes in either linear (weak) or saturated (strong) limits may be developed, with the fluorescence collected directly from the laser-excited species or indirectly from bath gases that are in vibrational resonance with the laser-excited species. An example of the latter scheme is shown in Fig. 2, where PLIF images are obtained of CO fuel and the CO/CO_2 interface of a laminar diffusion flame, by using an identical excitation of CO . Near-resonant vibrational energy transfer (VET) processes carry energy from the laser-pumped CO molecules into the CO_2 molecules such that CO_2 fluorescence is observed at the CO/CO_2 interface.

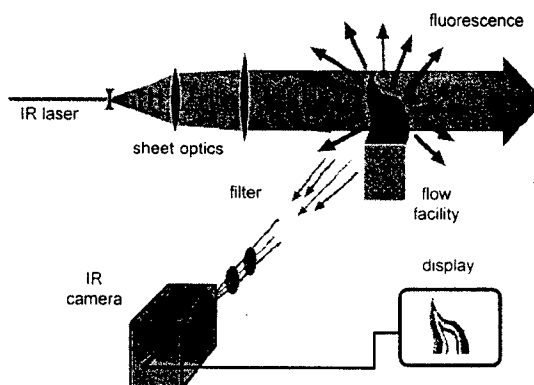


Fig. 1 Simplified Schematic of IR PLIF Imaging setup.

Linear excitation is appropriate for CO due to its slow vibrational relaxation. However, linear excitation is not well-suited for CO₂ imaging due to fast H₂O-enhanced VET and the attendant difficulty in interpreting the resulting signal. Saturated excitation using a CO₂ laser technique, however, has been found to be appropriate for CO₂, as it generates high signals and mitigates spatial variations in fluorescence quantum yield. Fig. 3 shows results from IR PLIF imaging of CO₂ in a lifted diffusion flame along with comparisons to benchmark CFD predictions and a composite of fifty, 5000-shot-averaged, 1-D, Raman measurements conducted at Yale by Smooke and Long.

Accomplishments in this work include the first IR PLIF diagnostics for CO and CO₂, the first detailed analysis of excitation schemes for these species, the first results using saturated laser excitation, and the first techniques that use combined laser / VET pumping schemes. These diagnostic developments enable single-shot images of these species in a manner not previously realized. Since IR PLIF is in principle applicable to all IR-active species, it has high potential for expanding the diagnostic possibilities available to combustion researchers, which might include visualization of the preflame (fuel) region of lifted flames, CO formation regions in flames, or mixing of hot exhaust gases with the atmosphere.

Ketone Photophysics for Quantitative PLIF Diagnostics

Our past research on acetone fluorescence included the following elements:

- 1) fundamental photophysical research to quantify the relative strength of fluorescence as a function of temperature, pressure, and bath gas environment, and
- 2) using the fundamental results, development and demonstration of PLIF techniques to measure relevant parameters in flowfields of practical interest (e.g. mixing regions of combustors).

To this end, we studied acetone fluorescence up to 1000 K and 16 atmospheres and developed a photophysical model that allows extrapolation beyond current experimental data. We also

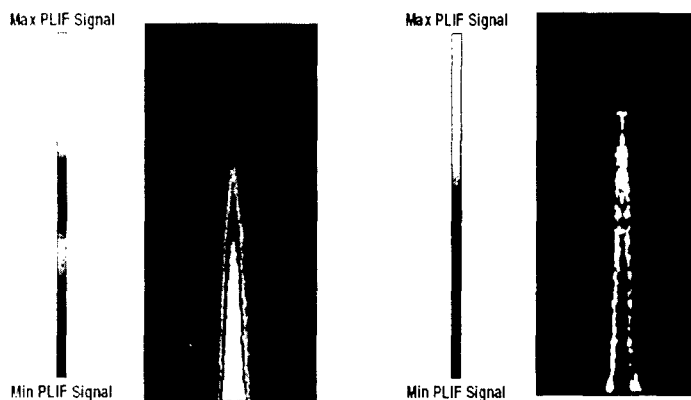


Fig. 2 The above two PLIF images of a steady CO/Ar/H₂ diffusion flame are realized by the excitation of the R(12) line of CO (2.35 μ m) followed by collection of CO fluorescence to visualize the fuel (left), and collection of CO₂ fluorescence to visualize the fuel/product interface (right).

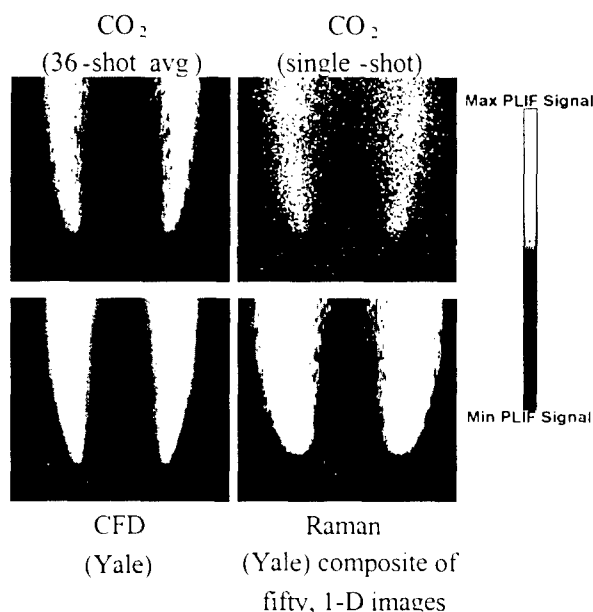


Fig. 3 Measurement and prediction of CO₂ distribution in a laminar lifted CH₄ diffusion flame. Clockwise from top left: 36-shot average IR PLIF image of CO₂; single-shot image of CO₂; overlay of fifty, 5000-shot-averaged, 1-D, Raman images (Yale); and computational prediction of CO₂ concentration (Yale). Yale results are courtesy of M. B. Long and M. D. Smooke.

demonstrated simultaneous, two-dimensional temperature and concentration measurements in unsteady flows.

Recently, we have begun to study attractive alternatives to acetone using the same fundamental approach. This was motivated by the need for a variety of tracers with physical properties (e.g. boiling points, diffusion coefficients) that match relevant fuels. Other ketones, because of their photophysical similarity to acetone (boiling point 56 °C) are primary candidates. We are thus investigating larger molecules like 3-pentanone (boiling point 100 °C), and we have discovered some key differences between pentanone and acetone. Most importantly, the ratio of fluorescence from two different excitation wavelengths as a function of temperature is not the same for acetone and 3-pentanone; rather, 3-pentanone tends to be more temperature-sensitive for similar excitation wavelengths (e.g., see Fig. 4, comparing the two 308/248 nm curves). The trade-off for this increased temperature sensitivity is a decrease in the signal at high temperatures. However, the temperature range of 3-pentanone PLIF can be extended by choosing a slightly less sensitive wavelength combination, e.g. 308/266 nm excitation. Through continued fundamental studies, a ketone database is being developed that will allow optimal selection of tracer and excitation strategy for a broad range of practical applications.

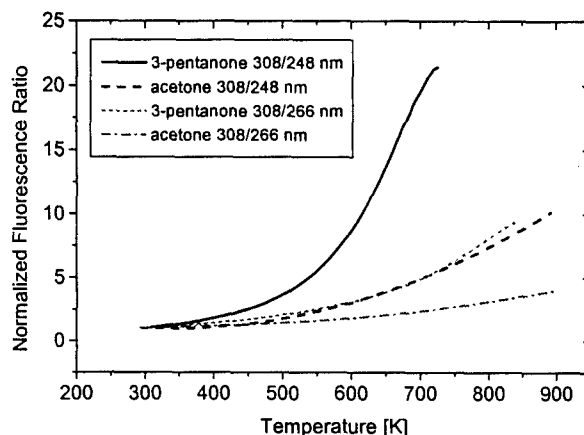


Fig. 4 Fluorescence ratio of acetone and pentanone as a function of temperature for two combinations of excitation wavelength.

Blue Diode Laser Absorption Sensor for NO₂

A highly sensitive and economical NO₂ diagnostic has been developed using a stable diode laser light source tuned to 390.130 nm. Single-pass absorption techniques were used to measure the absorption cross-section of NO₂ at 298 K in a flowing cell, and at elevated temperatures behind incident and reflected shocks in a shock tube. Optical absorption at this wavelength, in the dense NO₂ A-X spectrum, was found to have little variation with wavelength and pressure, thus simplifying analysis. The absorption cross-section, shown in Fig. 5, was found to vary approximately linearly with temperature. The utility of this diagnostic was demonstrated by studying NO₂ decomposition at temperatures ranging from 1500 K to 1900 K, and comparing the results with model predictions using established reaction rate constants.

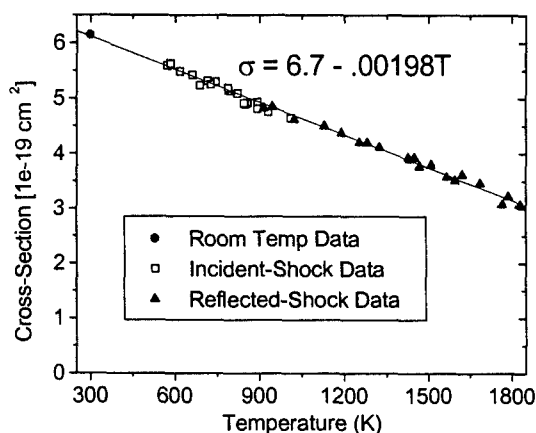


Fig. 5 NO₂ cross-section vs. temperature at 390.13 nm (at pressures of 0.2 to 1.7 atm).

Laser-Induced Fluorescence Diagnostics for NO in High-Pressure Combustion Gases

Laser-induced fluorescence (LIF) of NO in the A-X bands has become an important tool for monitoring pollutant production and for measuring properties such as gas temperature or velocity in flows seeded with trace quantities of NO. Earlier work at Stanford identified two difficulties which must be overcome for routine application of NO LIF at combustion temperatures and high

pressures. First, pressure broadening causes NO and hot vibrational O₂ transitions to overlap. As a result, LIF from O₂ obscures the NO signal, especially in fuel-lean, high-pressure environments found in advanced gas turbine combustors. Second, the spectroscopic database for NO and O₂ does not contain sufficient hot band positions, pressure-broadening information, or collisional energy transfer rates (rotational and vibrational) for quantitative LIF measurements.

NO (X²Π) has three vibrational levels with sufficient population at combustion temperatures for practical LIF: $v=0, 1$, and 2 . We began a systematic investigation of high-pressure NO LIF, emphasizing the A-X (0,1) transitions where there is a paucity of data. In a new high-pressure, premixed burner, we excited nascent and 300 ppm seeded NO using wavelengths between 236.5 and 237.5 nm at pressures between 1 and 60 bar. The ensuing fluorescence was dispersed in an imaging monochromator, and the fluorescence spectrum was recorded with an intensified CCD array. We observed that the NO structure is discrete at 1 bar, becomes blended by pressure broadening at 10 bar, and is dominated by O₂ interference at 40 bar. The NO rotational structure is not discernable at 60 bar. Work is in progress to quantify and model these spectra and to evaluate the relative merits of measuring NO by LIF of the A-X (0,0), (0,1) or (0,2) bands.

The choices between NO LIF strategies are complicated by unexpected optical absorption of hot flame gases; e.g., in-cylinder absorption spectra indicate that light at 225 nm (NO A-X (0,0)) is significantly absorbed by unknown species and bands. We have used a newly-developed UV kinetic spectrograph to measure the optical absorption of shock-heated CO₂ between 190 and 300 nm and found surprisingly strong temperature dependence in this optical absorption. This recent observation may explain the unexpected UV absorption of hot combustion gases.

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